

Remarks

Claims 6 - 14 are pending. Favorable reconsideration is respectfully requested.

The present invention is directed to the production of vinyl acetate. Vinyl acetate is a commodity chemical produced in large amounts. Hence, as with all high volume commodity chemicals whose processes of preparation are technologically mature, even very small increases in yield, throughput, reduction of reactant costs, minimization of byproducts, etc., are very important commercially, even though when viewed in isolation the improvement might seem small (i.e. <0.1% improvement).

In vinyl acetate production, ethylene, acetic acid, and oxygen react at elevated temperature in the gas phase in a fixed bed reactor. Ethylene conversion is very incomplete, as is also acetic acid conversion, and thus these components must be recycled to the process if economic operation is to be expected. Byproducts of the reaction include relatively large amounts of carbon dioxide as well as methyl acetate and ethyl acetate. In addition, large amounts of "inerts" (inert gases) accumulate, which must be removed from the system. If not removed, the inerts lower vinyl acetate selectivity. The inerts are supplied in numerous sources, for example in the oxygen feed (argon, nitrogen), with the acetic acid (nitrogen) and with the ethylene (methane, ethane).

Due to the low conversion of ethylene and its relatively high cost, the ethylene must be recycled. However, since the product stream contains numerous other substances besides vinyl acetate product and ethylene, including carbon dioxide and inerts, simple recycle has not been possible.

Russell GB 1 264 377 ("*Russell*") is directed to a process for vinyl acetate production with increased conversion of ethylene. *Russell* was filed in 1968, forty years ago, and attests to the long established need for maximizing ethylene usage. Prior to *Russell*, as discussed in *Russell* in column 2 of page 1, ethylene and acetic acid were previously recycled back to the

reactor. However, *Russell* indicates that this recycle causes disadvantages. Hence, *Russell* teaches avoiding total recycle of the ethylene-containing product stream. Instead, *Russell* divides the product stream into a recycle stream and a "purge" stream. The recycle stream, containing ethylene, carbon dioxide, and inerts, is recycled back to the initial reactor. The purge stream is directed to a second reactor for vinyl acetate production. The method of *Russell* has several important drawbacks. First, for complete utilization of ethylene, more than two reactors are needed, since the conversion of ethylene in each reactor is only about 10%. Second, the purge reactor effluent, which still contains ethylene and acetic acid, is discarded, resulting in the loss of valuable raw materials. Finally, the recycle stream to the first reactor cause an undesirable build-up of inerts which cause a reduction in selectivity in the main reactor. *Russell* was able to achieve an increase in overall yield of vinyl acetate, but at high cost.

That the invention of *Russell* still required improvement is illustrated by Williams et al. published European application EP 0 985 657 ("*Williams*"). In *Williams*, the problems of inerts accumulating in the main reactor are thoroughly discussed on page 4. *Williams* improves upon the prior art by increasing the mol% of ethylene in the reactor to above 60 mol%. *Russell*, for example, used only about 10 mol%. However, *Williams* indicates that due to the high ethylene concentration, inerts are a much more serious problem. While inerts were tolerable (with decreased selectivity) in *Russell*, they are intolerable in the *Williams'* process.

In *Russell*, for example, inerts, principally nitrogen, constituted about 75 volume percent (and hence roughly 75 mol% also) of the principle reactor contents. Thus, while recycle of a high inerts content recycle stream back to the reactor will increase the inerts content, it does so only slowly, since the percent inerts in the reactor is already very high¹ and the recycle stream is similar (75% N₂ main reactor feed, 77% recycle stream).

In *Williams*, the ethylene content is much higher, 60 - 85%, and the reactor also contains oxygen and acetic acid. Therefore, the recycle of inerts will cause a serious dilution of

¹This is very dated technology. No one would remotely consider a process such as this, with its low mol% of ethylene, today.

the ethylene content. Thus, *Williams* indicates that numerous methods should be used to minimize the amounts of inerts initially introduced, but still requires venting of purge gas to minimize buildup of inerts and CO₂. Venting (with use of a flare) is very wasteful.

To maximize use of ethylene, *Williams* removes vinyl acetate and acetic acid from the process gas by condensation, removes CO₂ by potash scrubbing, and then separates ethylene from the remaining gases through use of a gas permeable membrane. All the ethylene is then recycled back to the reactor. Inerts must be carefully removed in order to maintain the high mol% of ethylene in the reactor.

In Applicants' invention, a high ethylene content is used as was used in *Williams*. Thus, the presence of inerts is an important factor. However, purification of recycle gas as taught by *Williams* requires a membrane separator, which is expensive. Applicants solved the ethylene recycle problem by first removing vinyl acetate from the product gas stream in a scrubber charged with acetic acid, removing CO₂ from the recycle gas, and recycling from 75 to 99% of the crude recycle gas back to the reactor, while directing 1 to 25% of the crude recycle gas to processes other than vinyl acetate production. Preferably, the amount of recycle stream recycled to the other process is 5 to 20%.

The Examples illustrate the unexpected and surprising results of the subject invention. The percentage of N₂ and CO₂ in the recycle stream are minimized to about 10% by volume, amounts which do not increase over time, thus requiring no wasteful purge of the reactor. When 15% diversion of ethylene is employed, the selectivity rises to 93%. By way of contrast, even at very high ethylene mol%, and purification of ethylene recycle², the selectivity of *Williams* process never rose above 90.8%. It is most surprising that the partial recycle of the crude product gas results in such a high selectivity. The prior art, e.g. *Williams*, teaches that the opposite should occur.

²*Williams*' examples actually used pure "virgin" ethylene.

Moreover, the ethylene diverted by Applicants is not wasted, but is directed to other processes consuming ethylene so its value is recovered. Note that the ethylene being diverted has been treated to remove CO₂, but not inerts or other contaminants. Prior to Applicants' invention, it was not recognized that such a "crude" source of ethylene could be used for other purposes, and it was merely purged, generally as a "flare" or burned for its fuel value (a very expensive fuel).

Claims 6 and 9 - 11 have been rejected under 35 U.S.C. § 103(a) over *Williams*. Applicants respectfully traverse this rejection.

As previously discussed, *Williams* recycles all the ethylene in the product gas stream back to the reactor, and is very careful to attempt to remove all inerts prior to doing so. Yet even with membrane purification of the ethylene, *Williams* still occasionally requires purging gas from the system. This is because, like Applicants, *Williams* operates at high ethylene concentration, *See, Williams*, page 4 at [0028] and the extensive list of methods to minimize inerts on page 4, lines 16 - 27. The entire thrust of *Williams* is to operate at very high ethylene concentration which requires minimizing inerts. *Williams* therefore does not teach or suggest that the ethylene product stream may be recycled without removing inerts, or that only a portion of this stream is recycled. *Williams* also does not teach or suggest that the ethylene not recycled be used in another process which consumes ethylene. *Williams* only teaches ([0028]) venting gas from the reactor to remove carbon dioxide and inerts. Since the reactor contains about 60 - 80 volume percent of ethylene, this venting, even though it may only occur periodically, is very wasteful. If it were obvious to direct a crude ethylene stream to another ethylene consuming process, *Williams* would certainly have mentioned directly a purge stream from the reactor to such a process. However, he does not.

Moreover, the only ethylene vented or purged from the *Williams* process is the vent stream from the fluid bed reactor. The ethylene stream of Applicants, still containing inerts, is not taken from reactor, but is taken from the system following removal of vinyl acetate by scrubbing with acetic acid, and removal of CO₂. *Williams* does not teach or suggest removing

a portion of inerts-containing ethylene from this source. Rather, he teaches full recycle of purified ethylene. In view of the substantial efforts of *Williams* to remove inerts from the ethylene recycle stream, it is most surprising that in Applicants' process, where the recycled ethylene contains inerts (claim 1: "without further non-reactive gases being separated"), the selectivity is even higher than that of *Williams*. Both systems employ a Pd/Au supported catalyst. Since inerts are known to reduce selectivity and space time yield in vinyl acetate processes employing high ethylene content, it would not be obvious to recycle an inerts-containing ethylene product stream back to the reactor. Thus, Applicants respectfully submit that the claims are non-obvious over *Williams*, and respectfully request withdrawal of the rejection over *Williams* under 35 U.S.C. § 103(a).

Claims 6 and 9 - 11 have also been rejected over *Russell* under 35 U.S.C. § 103(a). Applicants also respectfully traverse this rejection. *Russell* teaches use of a second reactor to further react ethylene from the product stream of the first reactor. The claims of the present invention require that the recycle of inerts-containing ethylene be back to the same reactor. Use of a second reactor is expensive. *Russell* also does not teach or suggest removal of CO₂ from his first (or second) reactors. Rather, all of the product stream used by *Russell* is used without separation of CO₂. The *Russell* process is a long outdated process not relevant to modern vinyl acetate production. Note *Russell* at page 3, column 2, lines 113 - 119, where he indicates that with volatile olefins such as ethylene, the olefin content of the reactor feed is preferably maintained at such a low level that olefin recovery from the plant purge is not economical. Applicants' process operates at a much higher ethylene content.

Like *Williams*, *Russell* does not teach or suggest that any ethylene be routed to a different process which consumes ethylene. Rather, all the ethylene in *Russell's* process is either recycled or purged and discarded. Moreover, the ethylene in *Russell's* process is not likely to be suitable for routing to other ethylene-consuming processes, since in addition to inerts, it also contains substantial amounts of CO₂. Withdrawal of the rejection of claims over *Russell* under 35 U.S.C. is respectfully solicited.

Applicants understand that the rejections just discussed are over *Williams* and *Russell* separately. If the Office meant the rejection to be over the combination of *Russell* and *Williams*, Applicants offer these further comments.

Russell and *Williams* are not properly combinable. In the case of *In re Anita Dembiczak* and *Benson Zinbarg*, 50 U.S.P.Q.2d 1614 (Fed. Cir. 1999), the CAFC has indicated that the requirement for showing the teaching or motivation to combine references is "rigorous." *Dembiczak* at 1617. Moreover, this showing, which is rigorously required, must be "clear and particular." *Dembiczak* at 1617. See also, *C.R. Bard v. M3 Sys., Inc.*, 48 U.S.P.Q.2d 1225, 1232 (Fed. Cir. 1998). It is well established that merely because references can be combined, the mere suitability for logical combination does not provide motivation for the combination. See, *Berghauser v. Dann, Comr. Pats.*, 204 U.S.P.Q. 398 (DCDC 1979); *ACS Hospital Systems, Inc. v. Montefiore Hospital*, 221 U.S.P.Q. 929 (Fed. Cir. 1984). Moreover, mere conclusory statements supporting the proposed combination, standing alone are not "evidence". *McElmurry v. Arkansas Power & Light Co.*, 27 U.S.P.Q.2d 1129, 1131 (Fed. Cir. 1993). See also, *In re Lee*, 61 U.S.P.Q. 2d 1430 (Fed. Cir. 2002).

Here, *Russell* "recycles" an ethylene product stream containing inerts and CO₂ to a second reactor. *Russell*, in the case of ethylene, also operates at low ethylene concentration. *Williams*, on the other hand, operates at high ethylene concentration, recycles all the ethylene back to the same reactor, and prior to doing so, removes inerts and CO₂ in order to minimize the inerts content in the reactor. These processes are completely different and cannot be combined.

Moreover, the law is clear that when references are combined, all the salient teachings of each reference must be contained in the proposed combination. See, e.g. *In re Wesslau*, 147 USPQ 391, 393 (CCPA 1965). This does not mean that every minutia must be contained in the combination, but it does mean that all salient features must be. The salient features of *Russell* are the use of a second reactor to further react an ethylene product stream, and the ability to do so without purifying the first reactor ethylene stream. Therefore, any combination of these references must include two reactors, with at least a portion of the first

reactor ethylene stream going to this second reactor. However, the claims require that ethylene recycle, having removed CO₂, go back to the reactor, not into a second reactor. Thus, the claims are also non-obvious over the combination of *Williams* and *Russell* if that rejection was intended. However, in view of the major differences in technology, Applicants do not believe that these references would be viewed as combinable by one skilled in the art, as stated previously.

Claims 6, 7, and 8 have been rejected under 35 U.S.C. § 103(a) over *Williams* and *Russell* further in view of numerous secondary references which have been cited as disclosing processes where ethylene is consumed. It is noted that the *Cooley* reference does not disclose ethylene at all. *Cooley* is directed to producing 1,4-butanediol from acetylene, not ethylene. Ethylene cannot be used in a Reppe reaction. Further, the *Baba* reference is a vinyl acetate production process. The claims require the inerts-containing the ethylene to be directed to an ethylene-consuming process other than a vinyl acetate process.

Moreover, the ethylene removed from Applicants' reactor process stream has been treated to remove CO₂, but not other non-reactive gases (inerts). One skilled in the art, prior to applicants' invention, would not be motivated to employ the inerts-containing ethylene in another process due to its high impurity level. *See, e.g.* the typical make-up of the gas after CO₂ scrubbing, which contains only about 80 - 83% ethylene. If such a stream were viewed by the art as being useful in other ethylene consuming processes, then *Williams*, *Russell*, or someone would have suggested this long ago.

Thus, neither *Russell* nor *Williams* nor their combination teach using the inerts-containing ethylene in another ethylene-consuming process, and neither do the secondary references. All the secondary references (except *Cooley* and *Baba*) teach only the use of ethylene, but are completely silent regarding the ethylene source.

Finally, neither *Williams* nor *Russell* nor their combination teach the remaining aspects of the subject invention claims, and thus even were the secondary references somehow construed to teach using an impure vinyl acetate ethylene product stream in another process,

since the remaining limitations are not taught or suggested, claims 6, 7, 9 are non-obvious over the combination of references proposed.

New claims 12 - 14 have been added to more particularly point out and distinctly claim preferred embodiments of Applicants' invention. Support may be found on page 5 of the specification.

The corresponding EP application has been examined and granted. The *Russell* and *Williams* references were considered by the EPO and claims corresponding to claims 6, 7, and 8 of the present application were allowed. Although Applicants' attorney is aware that the dispositions of foreign offices are not binding on the U.S. Patent Office, the similarity of the European and U.S. Patent laws with respect to obviousness (lack of invention) and novelty are persuasive evidence of patentability, especially when the same references are involved.

Applicants submit that the claims are now in condition for Allowance, and respectfully request a Notice to that effect. If the Examiner believes that further discussion will advance the prosecution of the Application, the Examiner is highly encouraged to telephone Applicants' attorney at the number given below.

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Respectfully submitted,

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